Correlating growth conditions with photoluminescence and lasing properties of mid-IR antimonide type II "W" structures

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We explored the evolution of the photoluminescence (PL) properties versus molecular beam epitaxy growth conditions for a series of type II "W" quantum well [InAs/GaInSb/InAs/AlAsSb] structures. The highest PL intensities are obtained when the quantum wells are grown in a temperature range between 487 and 507 °C. Cross-sectional scanning tunneling microscopy was used to explain the temperature evolution of the PL. AlAs clustering within the AlAsSb barrier was observed at low growth temperature. The PL intensity decrease at high temperature was related to In clustering in the GaInSb layer. Laser structures grown at both 425 and 500 °C displayed lower lasing thresholds, lower internal losses, and longer Shockley–Read lifetimes than any similar structures grown previously at NRL. A thicker optical cladding layer of 3.5 μm suppressed mode leakage into the substrate and reduced the internal loss to 2.1 cm^{-1} at 78 K. [DOI: 10.1116/1.1688805]

I. INTRODUCTION

The merits of the type II "W" quantum well (QW) structure for mid-IR semiconductor laser design are well known. These include strong electron-hole wave function overlap (despite spatial separation of the holes and electrons), good electrical confinement (due to large conduction and valence band offsets in the barrier layers), a two-dimensional density of states for both electron and holes (yielding higher differential gain), and suppression of the Auger nonradiative decay modes (leading to lower lasing thresholds at higher operating temperature).[1] A typical "W" period[2] consists of a hole QW sandwiched by two electron QWs, which are in turn bounded by barrier layers (e.g., InAs, GaInSb, InAs, AlAsSb). Optically pumped "W" structures have displayed higher pulsed[3] and continuous wave (cw)[4] operating temperatures than any other interband III–V lasers emitting beyond 3 μm. Electrically pumped lasers have operated in pulsed mode at room temperature[5] and cw up to 200 K.[6]

Despite these results, there have been few systematic attempts to optimize the growth conditions for "W" lasers,[7,8] or to correlate the various growth parameters with optical performance.[9] With this in mind, we have undertaken a systematic investigation of the effect of QW growth temperature, T_{active}, varied from 435 to 526 °C, on the photoluminescence (PL) for a series of InAs/GaInSb/InAs/AlAsSb structures. Such a study can be quite challenging, owing not only to the relative immaturity of antimonide molecular beam epitaxy (MBE),[10] but also to specific complexities associated with the "W" structure. In particular: (1) The interface chemistry is defined by a change of both cation and anion, leading to an additional interface bond-type degree of freedom in the MBE growth.[11] (2) The active region contains three distinct materials (InAs, Ga(In)Sb, and Al(As)Sb), whose ideal growth temperatures differ by more than 100 °C.[7] (3) The barrier layers are comprised of a mixed-group-V alloy which, owing to the sensitive dependence of group V incorporation rate on MBE parameters,[10] can be difficult to grow reproducibly. For this study, we have chosen to address these complexities in the following ways. At the interfaces no special shutter sequence was used, allowing the interplay between interface chemistry and thermodynamics to determine the interface bond type. Modulated-beam MBE growth[12,13] was used to generate the barrier layers. In this technique a random alloy is approximated by a suitably grown small-period superlattice, so that the group V ratio is primarily determined by shutter timing and layer thicknesses rather than the details of the growth environment. Finally, the growth temperature of a series of samples was varied by ~100 °C in order to encompass the relevant temperatures.

We also report on two optically pumped "W" laser samples grown at different T_{active} (425 and 500 °C) and with two different cladding layer thicknesses (2.5 and 3.5 μm). The first of these lasers was from a series of samples investigated recently to cross correlate the growth with various figures of merit.[14] In that study, it was shown that those wafers emitting more intense PL at 78 K nearly always yielded stronger PL at 300 K, narrower PL linewidths, narrower lasing spectral linewidths, lower lasing thresholds, and higher lasing efficiencies. Such observations imply that measuring the PL from simple test structures at 78 K provides a reliable tool for optimizing the QW growth. This procedure has led to the growth of lasers with optical figures of merit that are substantially improved when compared to previous type II "W" structures grown at NRL.

II. MBE GROWTH

The "W" structures described here were grown on a Riber 32P solid-source MBE machine equipped with valved

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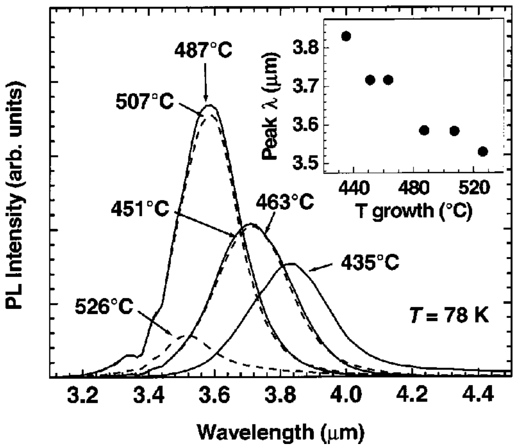

FIG. 1. PL spectra at 78 K for "W" structures with identical five-period active regions [$InAs(18\,Å)/Ga_{0.7}In_{0.3}Sb(34\,Å)/InAs(18\,Å)/AlAs_{0.106}Sb_{0.894}(234\,Å)$] and identical growth conditions apart from the indicated substrate temperatures. Inset shows peak wavelength as a function of growth temperature.

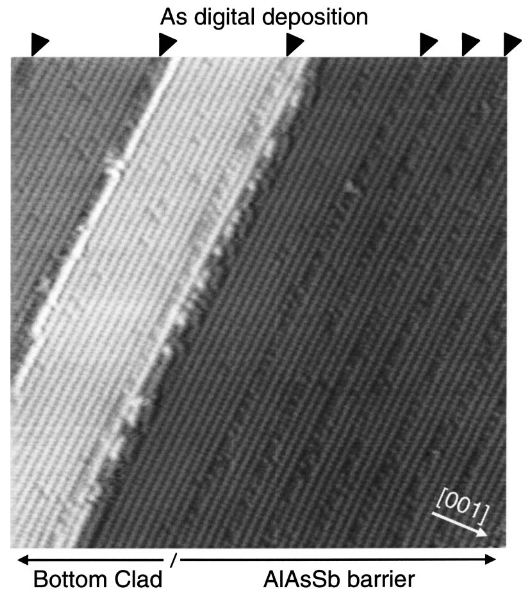

FIG. 2. Filled states XSTM image of the interface between the $AlAs_{0.08}Sb_{0.92}$ bottom clad and the first $AlAs_{0.106}Sb_{0.894}$ barrier, which were grown at 535 and 526 °C, respectively. The size of the image is 34 nm×34 nm, and has a gradient component added to the gray scale to accent the atoms on different terraces (sample bias −2.9 V, tunneling current 0.09 nA).

As and Sb crackers, except for one laser sample which was grown on a new Riber compact 21T system. T_{active} was measured using an optical pyrometer referenced to the known temperature for the GaSb (1×5) to (1×3) surface reconstruction (~429 °C for an Sb_2 rate of 2 ML/s).[15] An initial smoothing layer of 300 nm GaSb was grown at 500 °C (using epiready Te–GaSb wafers) followed by a lattice-matched $AlAs_{0.08}Sb_{0.92}$ buffer or optical cladding layer grown using modulated beam growth at a temperature of 540±10 °C. This layer (~0.5 μm thick for PL samples) was grown by alternately modulating the As and Sb shutters while keeping the Al shutter open. Next the "W" QWs were grown. For both PL and laser samples, each strain-balanced period consisted of the sequence $InAs(18\,Å)/Ga_{0.7}In_{0.3}Sb(34\,Å)/InAs(18\,Å)/AlAs_{0.106}Sb_{0.894}(234\,Å)$. PL samples incorporated five active "W" periods, while laser samples contained ten periods. These active periods were designed for emission in the 3.6–3.8 μm range at 78 K. For PL samples, all MBE parameters were held fixed during the QW growth except the temperature T_{active}, which varied between 435 and 526 °C. The growth rate was 1 ML/s except for the InAs QWs, which were grown at 0.3 ML/s. The structures were concluded with 15 nm GaSb for the PL samples and 500 nm GaSb for the laser samples.

III. PHOTOLUMINESCENCE AND XSTM CHARACTERIZATION

After deposition, the structural properties of the "W" wafers were characterized using high-resolution x-ray diffraction and Nomarski phase contrast microscopy. Those results (discussed elsewhere)[14] revealed high structural quality and low surface defect densities for all samples. Here we have further characterized the "W" wafers using cross-sectional scanning tunneling microscopy (XSTM), in order to identify atomic-scale variations in structure and composition from sample to sample.[16]

Figure 1 illustrates spectra at 78 K for the set of PL "W" wafers with T_{active} varying from 435 to 526 °C. Details of the measurement were presented elsewhere.[14] The strong sensitivity of the PL intensity to T_{active} is not surprising since most earlier studies at both NRL and elsewhere showed a relatively narrow window of 400–450 °C for optimal "W" structure growth.[7,8,17,18] However, for the present samples we observe a systematic increase in the PL intensity as T_{active} increases from 435 to 507 °C, followed by a precipitous drop-off at the highest temperature of 526 °C. These results identify an optimal window for "W"-structure growth of 487–507 °C. Similar trends were observed in two other series of "W" wafers with identical test structures. In addition, there appears a systematic decrease in the peak emission wavelength as a function of growth temperature (inset to Fig. 1). This may be attributed to either a slow decrease in the indium sticking coefficient as the growth temperature is increased or smearing of the QW interfaces at higher temperatures, both of which lead to a reduction in the thickness of the InAs QWs.

We present an explanation of the PL intensity as a function of sample growth temperature based on the structural quality of the AlAsSb and GaInSb layers grown at different temperatures. Figure 2 shows an XSTM image revealing a cross section (i.e., As, Sb) of the anion sublattice through the interface between the bottom clad and first AlAsSb barrier. Note the presence of multiple steps on the (110) surface, which resulted from an imperfect cleave. Within each terrace, As atoms exposed by the cleave appear "darker" due to the shorter Al–As bond length.[16,19] As can be seen in Fig. 2, the digitally deposited As in the bottom clad is very well confined. The As, deposited digitally at 535 °C, appears to exist mainly as a digital AlAs layer with minimal As incor-

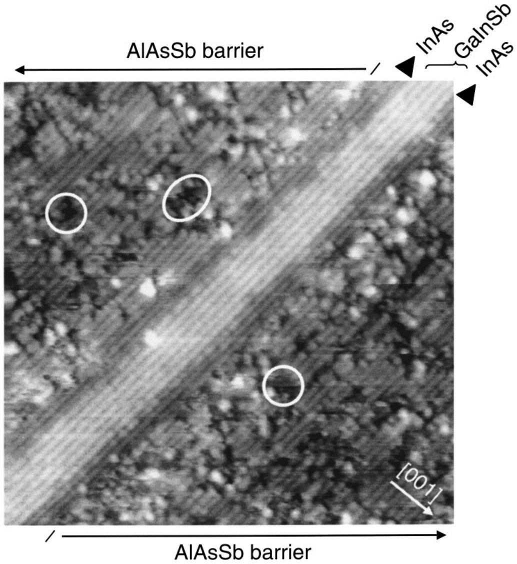

FIG. 3. Filled state XSTM image, 34 nm×34 nm, of an AlAsSb/InAs/GaInSb/InAs/AlAsSb active region grown at 435 °C. AlAsSb clusters are marked by white circles. The sample bias and tunneling current were −2.0 V and 0.07 nA, respectively.

poration within the AlSb. The excellent definition of the digital layers indicates that under these growth conditions the AlSb growth front is very flat (step flow) and that the digital AlAs layers are thermodynamically stable. We attribute the presence of a few isolated As within the AlSb layers to incorporation from the As background pressure. After the growth of the AlAsSb bottom clad the temperature was decreased slightly to 526 °C for the QW growth. It is clear from the XSTM image that at this slightly lower temperature the As is still confined relatively uniformly within 1–2 growth layers.

With decreasing T_{active} we observe a distinct change in the structure of the AlAsSb barrier layers. Small clusters of AlAs appear in the vicinity of the As digital layer (not shown). As the growth temperature decreases further, these clusters become larger as seen in the XSTM image of an active InAs/GaInSb/InAs/AlAsSb region grown at 435 °C (Fig. 3). Some of the AlAs clusters are so large they penetrate the 8 ML AlSb layer and extend into the AlAs layer beneath.

The observed AlAs clustering can be attributed, in part, to the evolution of the AlSb morphology as the growth temperature is varied.[20,21] As the temperature is lowered, the AlSb growth surface morphology becomes progressively rougher, with multilayer islands. For example, at 400 °C the growth front typically includes three or more AlSb layers. It appears that at these lower growth temperatures the As preferentially deposits at terrace edges and accumulates within vacancy islands (i.e., within the morphological "valleys") on the rough AlSb surface. Therefore, instead of a uniform digital AlAs layer, multilayer, nanoscale AlAs clusters are created. It should be noted that the earlier NRL studies which showed a preference for a lower growth temperature employed AlSb rather than AlAsSb barrier layers.[7,8] Also, lasers grown at the Air Force Research Laboratory (AFRL) did not contain Al at all,[17] and so the optimal growth conditions may have been different.

So far we have described a process whereby the morphology of the AlAsSb barrier layer improves with increasing growth temperature, resulting in a concurrent increase in PL intensity. However, the observed PL intensity goes through a maximum at 487–507 °C. This indicates that there must be at least one other process responsible for the change in PL behavior at higher growth temperatures; we suggest that the PL degradation is associated with In clustering in the GaInSb hole well of the "W" structure.

In earlier XSTM studies of "W" structures,[22,23] noticeable In clustering was observed within GaInSb layers grown at ~500 °C, with a typical cluster size of 5–10 nm. However, in structures grown at 435 °C little clustering was observed. In our current "W" structures, we do not observe In clustering following growth at 435 or 463 °C. We have been unsuccessful, to date, at obtaining atomic-resolution XSTM results for higher temperature samples because they usually cleave with large step structures along the GaInSb layer. However, we speculate that these cleavage steps may be related to compositional inhomogeneity, such as In clustering, which degrades the PL of the QWs.

IV. LASER CHARACTERIZATION

We next present lasing data for the two "W" wafers, which were grown with contrasting T_{active} and bottom clad thickness. These lasers were pumped with 100 ns pulses from a 2.1 μm Ho: yttrium–aluminum–garnet laser focused to a stripe of 150 μm full width at half maximum. We first note that the PL intensity for the laser grown at 500 °C ("21T") was a factor of 2 greater than that for the sample grown at 425 °C ("32P"). This is consistent with the PL data for the former, because T_{active} falls within the optimal growth window of 487–507 °C. Second, at 78 K the two lasers had very similar threshold pump intensities ($I_{th}=170\,W/cm^2$). This observation is *inconsistent* with our earlier findings that the lasing thresholds exhibited a strong correlation with PL intensity.[14] It is not clear why the 21T sample does not support this trend. One would expect that at 78 K the Shockley–Read nonradiative lifetime should govern both the PL intensity and the lasing threshold.

Lasing spectra for both the 21T and 32P lasers at three different temperatures are shown in Fig. 4. The multiplicity of spectral peaks in the laser with the thinner clad (2.5 μm) was identified in an earlier publication[24] as spectral modulation caused by leakage of the guided mode into the substrate.[25,26] A calculated modulation period of 25–30 nm calculated for the relevant substrate thicknesses is consistent with the observed spectra shown in Fig. 4. In addition, an associated increase of the internal loss, α_{int} (and subsequent reduction in the quantum efficiency), was predicted. We have verified this assertion with the growth of the 21T laser with 3.5 μm clad thickness. Note the much cleaner spectra, with nominally single lobes, at all temperatures in Fig. 4. In addition, the relatively broad spectra (40–150 nm) observed in the earlier growths have given way to narrower spectral

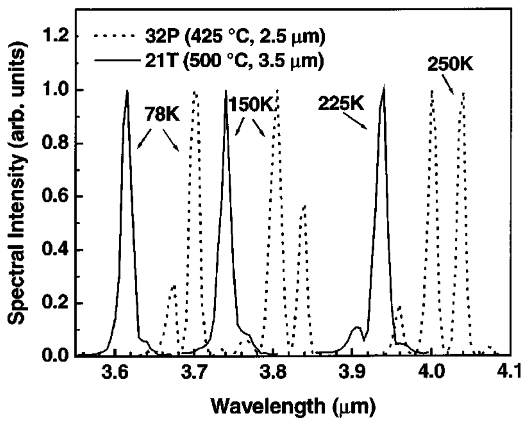

FIG. 4. Lasing spectra of three different temperatures for the 32P sample grown at 425 °C with a 2.5 μm cladding layer and the 21T sample grown at 500 °C with a 3.5 μm cladding layer. The observation of a broad spectral envelope containing multiple peaks is characteristic of all of the lasers grown with the thinner (2.5 μm) clads. However, the modulation has disappeared for the thicker (3.5 μm) clad.

widths of 15–35 nm. While this is a marked improvement, the linewidths are still surprisingly broad in light of the narrow PL. As long as the gain spectrum is smooth, theory projects that the lasing envelope should be no wider than a few nm at low temperatures where the carrier heating should not exceed ≈10 K. Sample inhomogeneities apparently cause the lasing linewidths to broaden somewhat.

We have quantified α_{int} by measuring the cavity-length dependence of the inverse efficiency[3,27] for a series of lasers from the 32P and 21T growths. The results as a function of temperature are shown in Fig. 5. For the 32P we derive $\alpha_{int}=4.5\,cm^{-1}$ at 78 K, which is consistent with a high reflectivity from the bottom of the substrate, whereas it is substantially lower than the loss expected due to substrate leakage in the absence of reflectivity feedback.[24] We see a further improvement for the 21T device, for which the losses of $\alpha_{int}=2.1\,cm^{-1}$ at 78 K and 46 cm^{-1} at 300 K are lower by more than a factor of 2. These results represent an even greater reduction from the best values in the 20–30 cm^{-1} range for earlier "W" lasers grown at NRL,[3] and also slightly lower than findings for a 10 QW optical-pumping injection cavity "W" structure grown at Sarnoff (e.g., $\alpha_{int}=5.5\,cm^{-1}$ at 78 K).[28]

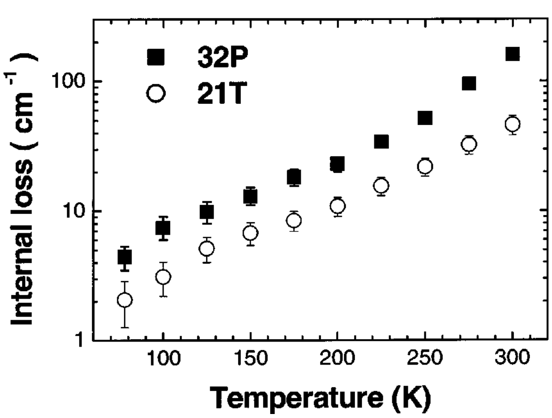

FIG. 5. Internal loss vs temperature, from analyses of the cavity-length dependencies of the inverse efficiencies for the 32P and 21T samples.

V. SUMMARY

We have explored the growth temperature evolution of the PL for type II "W" lasers using atomic-level characterization. It was found that whereas increasing the growth temperature improves the quality of the digital AlAsSb layer, it promotes clustering of the In within the GaInSb quantum well. We believe that the competition of these two processes results in an increasing PL intensity up to ~500 °C, where there is a maximum PL window, followed by a dramatic decrease at higher T_{active}. Improvement in the type II "W" laser performance was achieved by both increasing the QW growth temperature from 425 to 500 °C and incorporating a thicker bottom cladding layer (3.5 μm) to reduce the internal loss.

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